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Estimated Changes in Human Exposure to Suspended Sulfate Attributable to Equipping Light-Duty Motor Vehicles with Oxidation Catalysts

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The potential environmental impact of equipping vehicles with oxidation catalysts is estimated. Three independent techniques are used to appraise community exposures. Incremental increases in suspended sulfates and sulfuric acid aerosols are evaluated in terms of the number of vehicles equipped with oxidation catalysts.

In this paper, three independent techniques are used to estimate increased ("incremental") sulfate exposure which might result from the introduction of vehicles equipped with catalysts.

Sulfuric Acid-Suspended Sulfate Exposures

Conversion of fuel sulfur to suspended sulfates and sulfuric acid would not be expected to increase substantially the ambient levels of these pollutants over an entire air basin because the sulfur dioxide that is emitted from vehicles not equipped with catalysts is thought to be more or less completely converted to suspended sulfates through a series of complex chemical reactions in the atmosphere. On the other hand, conversion of fuel sulfur to suspended sulfates and sulfuric acid in the exhaust stream concentrates the sources of these pollutants in the breathing zone along arterial thoroughfares, in downtown street canyons, and in shopping centers.

Human exposures resulting from these locally increased concentrations can be estimated by a two-step procedure that considers ambient air quality and human activity patterns. Three independent methods have been used to estimate exposure.

As one approach, EPA used dispersion models developed for emissions of stable

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gases (carbon monoxide) from motor vehicles and ambient carbon monoxide (CO) data to estimate sulfate air quality changes. Estimates of the incremental sulfate exposure were then derived, by using the appropriate projected concentrations, for a designated behavior pattern.

A second estimate was made by using carboxyhemoglobin levels in nonsmokers living in major cities to estimate both existing carbon monoxide exposures and anticipated incremental sulfate exposures. This method, called the carboxyhemoglobin surrogate method, avoids assumptions about traffic patterns or human behavior but depends on an assumed relationship between carbon monoxide and suspended particulate sulfate emission rates. This model also assumes that the total CO exposure of nonsmokers can be attributed to emissions from motor vehicles and that a predictable relationship exists between ambient CO and blood carboxyhemoglobin concentrations.

A third technique used lead surrogate data to estimate short-term incremental sulfate concentrations.

Carbon Monoxide Dispersion and Lead Surrogate Estimates

The basic assumptions of the CO dispersion model are: (a) acid aerosols and fine particulate sulfates disperse like gases; (b) incremental sulfate emissions from catalyst-equipped vehicles are 0.05 g/mile; (c) 25% of vehicle miles traveled (VMT) or two model years with catalyst-equipped vehicles; (d) 1-hr commuter trip of 30 miles on a ten-lane expressway with traffic flow of 20,000 vehicles/phr. Typical and adverse meteorological conditions are considered. Additionally, estimates for exposure at work and at home are made. It should be noted that many of these assumptions are based on "best judgements" that may tend to either overestimate or underestimate the real world situation. For example, the hypothetical commuter trip may overestimate exposures, whereas the fact that the model takes no account of sulfate emissions from vehicles on nearby roadways may underestimate them. However, this model provides a reasonable prediction for severe conditions and locations which might arise after two model years are equipped with catalysts.

Projections of the CO dispersion model were compared with roadside measurements of particulate lead emitted by motor vehicles. Measurement of the ambient lead levels were assumed to have been conducted during the typical meteorological conditions. Even with this uncertainty, it is of interest that the lead values predicted by the dispersion model are in general agreement with the ambient lead values measured near Los Angeles freeways.

The lead data were also used as a surrogate model to estimate incremental sulfate concentrations on busy expressways after 25% VMT are in vehicles equipped with catalysts. An adjustment was made for the slightly lower emission factor assumed for sulfates (0.05 g/mile) compared to 0.07 g/mile for lead. This lead surrogate model may underestimate concentrations because of lead particles settling near the roadside, or overestimate the concentrations as a result of reentrainment of settled lead particles by the traffic stream. Projected sulfate concentrations from this method (Table 1) are in the general range of values predicted by the CO dispersion model.

Dispersion model and lead surrogate approaches were used to estimate short-term incremental ambient concentrations to which automobile passengers, pedestrians near expressways, pedestrians in urban street canyons or shopping centers, residents near expressways, and residents five blocks from the busiest expressways would be exposed (Table 2). The multiplying effect of urban street canyons on pollutants emitted from low level sources was estimated to result in concentrations three to five times the values that would be predicted by the normal dispersion model for a given traffic density. The effect of slow or stalled traffic on the emission factor (0.05 g/mile) for sulfates used

Table 1. Predicted short-term incremental exposures to suspended particulate sulfates and sulfuric acid emitted from vehicles equipped with oxidation catalysts. [a]

Human exposure	Highway source	Method of estimation	Incremental exposure, $\mu g/m^3$			
			Normal meteorology, wind directly across highway	Normal meteorology, wind at worst angle	Worst meteorology, wind directly across highway	Worst meteorology, wind, at worst angle
Automobile passengers	Expressway	Dispersion model	2	—	—	—
		Lead surrogate[b]	4–5	7	22	124
Pedestrians	Near expressway	Dispersion model	2	—	—	—
		Lead surrogate[b]	3–7	5	20	88
Pedestrians	Urban street[c] canyon[c]	Dispersion model	4–6	—	—	—
		Lead surrogate[b]	2–9	—	—	—
Nearby residences	Expressway	Dispersion model	1	2	19	33
Residences about five blocks away	Expressway	Dispersion model	0.3	0.3	5	6

[a] Emission assumptions: 25% of vehicle miles traveled on busiest multilane expressway (20,000 vehicles/hr at 30 mph by vehicles) equipped with oxidizing catalysts emitting 0.05 g of sulfuric acid of sulfate per vehicle mile. Other 75% of vehicle miles involve no emissions of sulfuric acid and suspended sulfate. Peak hourly values are tabled.

[b] Computed by multiplying observed lead levels by adjustments for differences in emissions per vehicle mile and percentage of vehicle miles emitting the particulate of interest: (observed lead level) × (0.72) × (0.25) or 0.18 times observed lead level for 3-hr average.

[c] Assumes 10,000 instead of 20,000 vehicles/hr. Actual observed carbon monoxide values in medium-sized suburban shopping center indicate that peak hourly exposures to 4.6–5.3 $\mu g/m^3$ of carbon monoxide and by inference 1–2 $\mu g/m^3$ suspended sulfates and acid aerosols would occur.

Table 2. Predicted incremental exposures to suspended particulate sulfate and sulfuric acid aerosols emitted from vehicles equipped with oxidation catalysts.

Exposure model	VMT travelled by catalyst-equipped vehicles, %	Incremental sulfate exposure (24 hr avg), $\mu g/m^3$	
		Typical	Adverse
Dispersion model	25	1	9
Hemoglobin surrogate	25	2	4
Dispersion model	100	5	35
Hemoglobin surrogate	100	9	15

in the CO dispersion model is not known. Meteorological conditions are usually less conducive to dispersion in the late evenings and early mornings than from late morning to evening. One might, therefore, expect that peak hourly levels of acid aerosols and suspended sulfates as well as their lead surrogate would be somewhat higher during the morning rush hours than the evening rush hours. This was observed in the ambient lead measurements in Los Angeles.

On assuming that two model years are equipped with catalysts, increases in ambient air concentration along the busiest expressways are estimated to range from 2 to 5 $\mu g/m^3$ for periods ranging from 1 to 3 hr/day under normal meteorological conditions, e.g., commuters on a busy expressway for an hour would have their exposure increased by 2–5 $\mu g/m^3$. Similarly, increases in ambient levels for 1–3 hr/day will range from 2 to 7 $\mu g/m^3$ for pedestrians near expressways and from 2 to 9 $\mu g/m^3$ for pedestrians in downtown street canyons and shopping centers. All of the previous values would quadruple if all cars were catalyst equipped. Exposures would increase as meteorological conditions become less favorable to dispersion.

Persons residing or working near busy expressways would be exposed to short-term increments of about 2 $\mu g/m^3$ under normal meteorological conditions. However, adverse meteorologic conditions would be expected to result in much higher incremental exposures. Under these conditions, which might be expected to occur on 1% of the days in each year, peak hourly concentrations outside of homes nearest the expressways could exceed 30 $\mu g/m^3$. When meteorological conditions are intermediate, the expected exposure increments would be less. For example, under conditions when the atmosphere is stable and when there are low wind speeds perpendicular to the highway, exposures would approach 20 $\mu g/m^3$ at homes nearest the expressway and 5 $\mu g/m^3$ at homes five blocks from the expressway.

Short-term peak exposures to suspended sulfates and acid aerosols lasting from less than 1 hr to several hours are probably sufficient to trigger asthmatic attacks or cause worsening of the symptoms that accompany chronic heart and lung disease. Repeated, short-term peak exposures may also be just as significant in inducing delayed adverse health effects as the more persistent but lower exposures lasting months or years. Our present scientific information base is not adequate to quantify either the acute or delayed effects of single or repeated short-term exposures to suspended sulfates or acid aerosols lasting less than 24 hr. There is, however, information linking 24-hr suspended sulfate exposures to an increased frequency of asthma attacks and aggravation of chronic heart and lung disease: exposures higher than 8–10 $\mu g/m^3$ for 24 hr have been demonstrated to produce these adverse effects (*1*). Using the carbon monoxide dispersion model, the incremental 24-hr sulfate exposure attributable to catalysts was estimated by examining a plausible daily behavior pattern. Commuters living adjacent to a busy expressway are assumed to spend about 2 hr daily driving along an expressway, 1 hr daily in an urban street canyon or shopping center, 8 hr at work, and 13 hr at home. When only two model years are equipped with catalysts, incremental daily exposures to these commuters could vary from 1 $\mu g/m^3$ on days with typical meteorological conditions to 9 $\mu g/m^3$ on days with adverse conditions (see Table 2). If all vehicles were equipped with catalysts, the 24-hr exposures would be 5 and 35 $\mu g/m^3$, respectively, for the two meteorological conditions considered.

Carboxyhemoglobin Surrogate Estimate

Blood carboxyhemoglobin levels can be used to estimate to CO exposure of a population. The resulting CO exposure estimates can be converted into incremental sulfate exposure estimates if all or most of the CO exposure of the population group under consideration is attributable to automobiles, and if the relationship between automotive emissions of sulfate and CO is known.

Carboxyhemoglobin levels in healthy blood donors living in 18 cities in the United States were measured by Stewart et al. (*2*) during 1970–72. Some of these data have been used in this analysis to estimate sulfate exposures. Los Angeles has relatively high ambient CO levels, which are largely attributable to automobile emissions. Therefore, the Los Angeles data were used to calculate the estimates discussed here.

Carboxyhemoglobin levels were converted to 8-hr CO exposures by using the methods of Coburn (*3*). Incremental sulfate exposures were then estimated with the assumptions that average CO emissions from the 1971 auto population were 74 g/mile and that catalyst-equipped cars emit 0.05 g/mile more sulfate than cars not catalyst-equipped.

The median and 95th percentiles of the Los Angeles carboxyhemoglobin measurements have been used to estimate sulfate exposures under typical and adverse conditions. The resulting 24 hr average incremental sulfate exposure estimates are shown in Table 1. Thus, after two model years of catalyst vehicles, typical 24-hr sulfate exposure might increase by 2 $\mu g/m^3$, while the increase in exposure would be 4 $\mu g/m^3$ under adverse conditions.

The results in Table 1 must be interpreted cautiously. The "typical" and "high" exposure estimates are accurate only to the extent

Table 3. Distribution of daily suspended particulate sulfate levels in selected American cities.

City	Year	Daily suspended particulate sulfur, $\mu g/m^3$											
		Minimum level	Frequency distribution									Maximum	Arithmetic average
			10	20	30	40	50	60	70	80	90		
Long Beach, Calif.	69	3.5	5.0	7.4	9.4	9.9	12.5	15.5	17.1	20.1	41.8	41.8	14.4
	70	2.0	3.7	5.4	6.0	7.4	8.9	9.8	10.8	11.1	14.0	15.6	8.6
Los Angeles, Calif.	69	2.4	3.8	4.9	6.8	7.4	8.3	12.2	16.4	18.7	19.6	25.2	11.2
	70	2.0	2.7	4.2	6.9	11.3	12.8	13.9	15.5	17.8	28.2	40.5	13.2
San Francisco, Calif.	69	2.4	3.1	3.8	4.7	4.8	5.6	6.0	6.2	8.7	13.8	17.0	6.7
	70	1.0	2.2	2.7	3.4	3.6	3.9	4.2	4.8	5.0	6.5	9.4	4.2
Denver, Colo.	68	1.8	2.6	2.8	3.6	3.7	4.5	4.7	5.2	5.4	6.6	8.6	4.5
	70	0.8	1.8	2.3	2.8	3.9	4.1	4.6	5.5	5.6	7.7	12.0	4.5
Chicago, Ill.	69	8.4	10.1	12.2	3.1	14.1	15.8	17.4	20.1	24.0	30.7	44.9	18.8
	70	4.5	5.8	7.5	1.1	12.3	13.1	17.6	19.1	19.7	25.8	29.6	14.8
Portland, Me	68	4.0	4.3	6.0	8.4	11.3	14.9	16.2	17.4	31.7	36.6	66.7	19.0
	70	6.5	8.3	9.3	10.9	12.8	16.7	18.9	21.8	23.3	27.0	34.1	17.0
New York City	69	5.0	5.8	8.3	11.3	11.8	15.1	17.1	21.3	33.4	39.3	57.2	19.1
	70	8.4	11.7	12.4	16.0	16.4	20.2	20.8	24.3	30.5	37.3	47.5	22.2
Charleston, W. Va.	69	4.9	8.4	8.6	9.3	13.7	16.5	28.5	34.2	38.7	57.5	73.3	25.8
	70	8.4	11.1	13.4	14.2	14.7	18.1	20.8	23.5	39.3	50.3	81.1	25.0
Washington, D.C.	68	5.6	8.1	9.1	11.6	13.9	14.6	16.3	19.1	19.8	23.9	29.3	15.4
	69	6.2	7.5	10.2	11.4	12.8	13.1	13.8	14.8	16.1	19.8	33.9	13.9

[a] From National Air Sampling Network data (*4*).

that the following assumptions are accurate: the individuals examined in the Los Angeles study received minimal exposure to CO from nonautomotive sources; observed carboxyhemoglobin concentrations were related to CO exposures as described above for commuters; the CO and sulfate emissions rates used are correct; and the atmospheric dispersion of sulfates is similar to that of CO. None of these assumptions is believed to be unreasonable. However, presently available data are not sufficient to make possible a precise verification of the assumptions. Thus, the estimates in Table 1 suggest, but do not prove, that: "typical" incremental daily exposures to sulfates may be in the neighborhood of 2 $\mu g/m^3$ after two model years of catalyst-equipped cars and 9 $\mu g/m^3$ if all cars are catalyst-equipped and measures are not taken to reduce sulfate emissions. Some people may receive incremental daily sulfate exposures in the neighborhood of 4 $\mu g/m^3$ after two model years of catalyst cars and 15 $\mu g/m^3$ if all cars are catalyst-equipped and no measures are taken to reduce sulfate emissions. Five percent of the population could experience exposures greater than those shown in Table 1.

Effect on Total Sulfate Exposure

When two model years are equipped with catalysts, the "adverse case" projections of incremental 24-hr sulfate exposures range from 4 $\mu g/m^3$ to 9 $\mu g/m^3$. These values are increased fourfold after all the vehicles on the roads are equipped with catalysts. In order to estimate the impact of these projected incremental exposures, one must recognize that significant urban levels are already seen in many of our urban centers (see Table 3). In many cases, sulfate concentrations exceed the level where it is our judgement that adverse health effects may be detected in susceptible individuals (8–10 $\mu g/m^3$, 24-hr average). The total exposure to sulfates after two years of vehicles equipped with catalysts may not be the exact sum of the projected increments and the ambient sulfate levels now measured (due to a lack of spatial coincidence of the measured data and the calculated exposures, the actual exposure could be more or less than the simple sum), the increment due to catalyst-equipped vehicles will clearly increase the total sulfate exposure level. In all probability this will significantly increase the number of days per year when a sizable segment of these urban populations would be exposed to a 24-hr average of more than 8–10 $\mu g/m^3$. If no program were introduced to lower the sulfur content of gasoline or to install other control devices to limit sulfate emissions, both models predict that, after ten model years are equipped with catalysts, the sulfate exposure attributable to motor vehicles alone would exceed the 8–10 $\mu g/m^3$ 24-hr average on a significant number of days per year. If the emission factor is lower than estimated by a factor of 10, no measurable adverse health effects would be expected from incremental exposure even with 100% of the vehicles equipped with catalysts. On the other hand, projected exposure levels could be higher in areas where gasoline sulfur content is significantly higher than the value assumed in estimating the sulfate emission factor for this model (the national average of 0.03% sulfur content). The emission factor increases as the sulfur content of the gasoline increases.

In our best judgment, adverse health effects among the most susceptible group of commuters traversing the busiest expressways will probably not be measurable after one model year is equipped with catalysts but are likely to appear by the end of the second model year. As more and more vehicles are equipped with catalysts, similar effects will probably be measurable in susceptible commuters traversing less busy arterial thoroughfares. According to our models, this situation would be predicted to occur after two to six model years. It must be emphasized that these projections may be significantly affected when more extensive quantitative sulfate emission data become available. The value for 24-hr exposure increments could be substantially higher or lower, directly related to the emission data obtained under a variety of operating conditions. The implications of the energy crisis as it pertains to catalyst related sulfate emissions as well as the possible effects of implementation of transportation control plans are also largely unknown.

EPA has weighed the established pollution abatement benefits which are the reduction of hydrocarbon and CO emissions and the gasoline economy benefits of the catalyst against the projected sulfate problem, with all the uncertainties noted above. It is EPA's best judgment that catalysts should not be banned and that the interim 1975 light-duty vehicle emission standards should be implemented as scheduled. However, because of the possibility of adverse health effects related to the incremental sulfate exposure in the future, EPA is implementing the following accelerated program: (a) ascertain the actual values for sulfate emissions from both the current automobile population as well as catalyst equipped vehicles under a variety of operating conditions; (b) develop better dispersion and exposure models; (c) conduct more extensive epidemiological, clinical and toxicological studies on the health effects of exposure to particulate sulfates and acid aerosols; and (d) concurrently with the above programs prepare contingency option plans to assure the protection of public health. These will include studies on the feasibility of the production and distribution of low sulfur gasoline and development of sulfate trap technology.

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